Incoherent Mollow triplet

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A counterpart of the Mollow triplet (luminescence lineshape of a two-level system under coherent excitation) is obtained for the case of incoherent excitation in a cavity. Its analytical expression, in excellent agreement with numerical results, pinpoints analogies and differences between the conventional resonance fluorescence spectrum and its cavity QED analogue under incoherent excitation.

Mollow [1] discovered a striking type of spectral shape in the resonance fluorescence problem, where an atom is irradiated by a strong laser beam. The celebrated *Mollow triplet* [2], that results from transitions between atomic states that are dressed by the coherent light field, has since been a testbed of nonlinear optics. It stands as one of the fundamental spectral shapes of light-matter interaction, maybe second only to the Rabi doublet. Although the Mollow triplet is rooted in quantum physics and bears much quantum features itself, it arises from a fully classical light field. Its Hamiltonian, in the rotating frame of the laser and at resonance, simply reads $H_L = \Omega_L(\sigma + \sigma^\dagger)$, with Ω_L^2 the laser intensity and σ the only quantum operator, namely, the two-level system annihilation operator. Including the spontaneous decay of the emitter, in the Lindblad form $\mathcal{L}_\sigma(\rho) = (2\sigma\rho\sigma^\dagger - \sigma^\dagger\sigma\rho - \rho\sigma^\dagger\sigma)$, leads to a master equation $\partial_t\rho = i[\rho, H_L] + \frac{\gamma_\sigma}{2}\mathcal{L}_\sigma(\rho)$ from which one obtains the famous Mollow triplet lineshape:

$$S_{\mathrm{coh}}(\omega) = \frac{\gamma_\sigma^2}{8\Omega_L^2 + \gamma_\sigma^2}\delta(\omega) + \frac{1}{2\pi}\frac{\frac{\gamma_\sigma}{2}}{\left(\frac{\gamma_\sigma}{2}\right)^2 + \omega^2} + \frac{\gamma_\sigma}{\pi}\frac{16\Omega_L^2 - \gamma_\sigma^2 - \omega^2}{\gamma_\sigma^4 + 4(\omega^2 - 4\Omega_L^2)^2 + \gamma_\sigma^2(5\omega^2 + 16\Omega_L^2)}\,. \quad (1)$$

It is composed of an elastic scattering peak, the Dirac δ function, and the triplet itself, with a central Lorentzian peak of full width at half maximum (FWHM) γ_σ and two satellite peaks at the symmetric positions $\pm\Re(R_L)$ with Mollow splitting $R_L = \sqrt{(2\Omega_L)^2 - (\gamma_\sigma/4)^2}$ and FWHMs $3\gamma_\sigma/2$. This structure was observed a long time ago with atoms [3] and more recently also in a variety of solid state systems [4–8], with, as befits the above description, coherent excitation.

In this text we consider a close counterpart of this fundamental system, where the light field is initially fully quantized, and becomes continuous as a result of an incoherent and continuous pumping that feeds the system with a very large number of photons. This situation is realized—as for quantization of the light field—in cavity QED [9], where quanta of a trapped standing waves (the photons) can be brought to interact with an isolated emitter, and—as for the incoherent pumping—with semiconductor microcavities [10], where excitations are continuously poured into the system with no external coherence fed in by a driving field. The role of the emitter is, in this case, played by a quantum dot placed in the antinode of the microcavity field. In the cavity QED version of the resonance fluorescence physics, the system is described by the Jaynes-Cummings Hamiltonian (still at resonance): $H = g(a^\dagger\sigma + a\sigma^\dagger)$ with the cavity mode also quantized through the boson operator a. Cavity decay γ_a as well as incoherent pumping P_σ are described like before with a master equation $\partial_t\rho = i[\rho, H] + \frac{\gamma_a}{2}\mathcal{L}_a(\rho) + \frac{P_\sigma}{2}\mathcal{L}_{\sigma^\dagger}(\rho)$ where ρ is now the density matrix for the combined two-level-emitter/cavity system. We assume that $\gamma_\sigma \ll P_\sigma$, so much so that we can neglect it for simplicity (it is already very small in a typical system and we consider the case of large pumping). We also assume the condition of strong-coupling, where $g \gg \gamma_a/4$. This regime was recently observed in the cavity emission as a transition from the Rabi doublet to a single lasing line [11]. Since this transition has been claimed remaining in strong-coupling, it results from climbing the Jaynes-Cummings ladder [12], and as such, is a successful realization of quantum nonlinearities in these systems. The importance of this breakthrough for microcavity QED is however hindered by such a simple manifestation, in particular since other mechanisms can also result in a similar behaviour of Rabi splitting collapse without entering the quantum nonlinear regime [13]. Quantum features are generally better observed when probing the quantum emitter, rather than the cavity, whose close connections with the classical oscillator tend to surface rapidly and dominate strongly. The theoretical description, which is straightforward in the low excitation regime even when solving the system exactly [12], becomes computationally demanding when the lasing regime is approached, but good approximations can be sought [14]. In this text, we consider this physics of the highly nonlinear regime of the microcavity-QED system, that is, Jaynes-Cummings physics under a strong incoherent pumping. We find that, in good systems by the standard of today, a new type of Mollow triplet is obtained in the direct QD emission spectrum. It is a close counterpart of the classical Mollow triplet where light is described by a classical field [1], whereas it is here described as numerous quanta of the cavity mode. The coherence is acquired

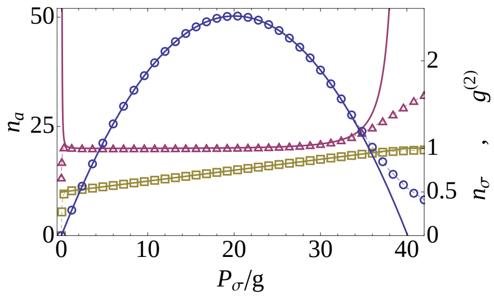

FIG. 1: (Color online) Exact numerical solution (points) and their analytical approximation, Eqs. (3), (lines), for n_a (blue/circles), n_σ (brown/squares) and $g^{(2)}$ (purple/triangles), for $\gamma_a = 0.1g$, as a function of pumping P_σ/g. The analytical solutions become unphysical when $P_\sigma = \kappa_\sigma$ (here at $40g$), where $n_a = 0$, $n_\sigma = 1$ and $g^{(2)}$ diverges. They are very good approximations in the region of interest, Eq. (4).

through the strong-coupling with the dot, resulting in striking variations from the case where it is provided by an external laser. We now describe them analytically.

Mollow regime — Whereas only one parameter (intensity) fully describes the light in Mollow's description, the Jaynes-Cummings picture requires from the start to take into account an infinite number of correlators between the fields, that we can however relate to each other [12]: $\langle a^{\dagger n} a^{n-1}\sigma\rangle = i\frac{\gamma_a}{2g}\langle a^{\dagger n}a^n\rangle$ and $\langle a^{\dagger n-1}a^{n-1}\sigma^\dagger\sigma\rangle = \big[P_\sigma\langle a^{\dagger n-1}a^{n-1}\rangle - \gamma_a\langle a^{\dagger n}a^n\rangle\big]/\big[P_\sigma + \gamma_a(n-1)\big]$ (all others being zero in the steady state). From this follows a first relation for the populations of the modes, $n_\sigma = \langle\sigma^\dagger\sigma\rangle$ and $n_a = \langle a^\dagger a\rangle$, namely $n_\sigma = (P_\sigma - \gamma_a n_a)/P_\sigma$. This also allows to obtain a self-contained equation for $\langle a^{\dagger n}a^n\rangle$:

$$\langle a^{\dagger n}a^n\rangle = \frac{\frac{nP_\sigma}{P_\sigma+(n-1)\gamma_a}\langle a^{\dagger n-1}a^{n-1}\rangle - \frac{2\gamma_a}{P_\sigma+n\gamma_a}\langle a^{\dagger n+1}a^{n+1}\rangle}{1+\frac{P_\sigma+(2n-1)\gamma_a}{\kappa_\sigma} - \frac{2P_\sigma}{P_\sigma+n\gamma_a} + \frac{n\gamma_a}{P_\sigma+(n-1)\gamma_a}}\,, \tag{2}$$

where $\kappa_\sigma = 4g^2/\gamma_a$ is the Purcell rate of transfer of population from the dot to the cavity mode. This recurrence equation allows to compute $\langle a^{\dagger n}a^n\rangle$ for all n as a function of n_a only. The solution for $n = 0$ gives a good approximation for the region where the cavity field behaves classically:

$$n_a \approx \frac{P_\sigma}{2\gamma_a}\Big(1 - \frac{P_\sigma - \gamma_a}{\kappa_\sigma}\Big) \quad \text{and} \quad n_\sigma \approx \frac{1}{2}\Big(1 + \frac{P_\sigma - \gamma_a}{\kappa_\sigma}\Big)\,. \tag{3}$$

The quality of this approximation is seen in Fig. 1 where it is compared with the exact solution, computed numerically [12]. The second order coherence function $g^{(2)}$ also admits a closed-form expression (not given here but plotted in Fig. 1) which is unity in good approximation. The expressions Eqs. (3) for the populations have a clear physical meaning, that can be discussed in terms of two parameters: the "*cavity feeding*", $F_a = P_\sigma/(2\gamma_a)$, and the "*dot feeding*", $F_\sigma = (P_\sigma - \gamma_a)/\kappa_\sigma$, efficiencies. At low pump, but still enough to be beyond the quantum regime [12], i.e., $\gamma_a < P_\sigma \ll \kappa_\sigma$, the cavity population increases linearly with pumping, with a half occupied dot. This is the most effective region for accumulation of photons in the cavity (the so-called *one-atom laser* [15]), with little disruption from incoherent processes. At smaller efficiency F_σ, the dot occupation also increases linearly with pumping, quenching the linear increase of the cavity population. F_σ represents, therefore, the degree to which the dot pumping succeeds in populating the dot itself, against the coherent exchange of population that feeds the cavity with efficiency F_a. These expressions are thus valid until the dot population is fully inverted, at $P_{\max} \approx \kappa_\sigma$, then, the self-quenching dominates the dynamics, emptying the cavity that goes to a thermal state. The maximum population of the cavity, $\max(n_a) \approx g^2/(2\gamma_a^2)$, is reached at the intermediate rate $P_\sigma \approx \kappa_\sigma/2$. This identifies the regime of interest for the observation of the Mollow triplet, where the cavity field is intense ($n_a \gg 1$) and coherent (with a Poissonian photon distribution, $T[n] = e^{-n_a}n_a^n/n!$ and $g^{(2)} = 1$):

$$\gamma_a \ll g < P_\sigma < \kappa_\sigma\,. \tag{4}$$

Now that we have a good and analytical description of the populations, we turn to the optical emission spectrum, that we show can be obtained in equally good approximations. The dot emission reads $n_\sigma\pi S_{\mathrm{inc}}(\omega) \equiv \Re\int_0^\infty\langle\sigma^\dagger(0)\sigma(\tau)\rangle e^{i\omega\tau}d\tau$. We compute the two-time correlator $\langle\sigma^\dagger(0)\sigma(\tau)\rangle$ in two steps: first, we solve the master equation in the steady state, finding the density matrix elements $\rho_{m,i;n,j}$ (for m, $n \in \mathbf{N}$ and i, $j \in \{0,1\}$, photon and exciton indexes, respectively). For the range of parameters of interest, we show that they can be analytically expressed only in terms of the photon distribution $T[n]$. Second, we apply the quantum regression formula.

1. Steady state density matrix — We consider only elements that are nonzero in the steady state: the populations $p_i[n] = \rho_{n,i;n,i}$ with $i = 0, 1$, corresponding to the probability to have n photons with (p_1) or without (p_0) exciton, and the off-diagonal terms $q_i[n] = \Im(\rho_{n,0;n-1,1})$, corresponding to the coherence between the states $|n,0\rangle$ and $|n-1,1\rangle$. The master equation now reads:

$$\partial_t p_0[n+1] = \mathcal{D}_{\mathrm{phot}}\{p_0[n+1]\} - P_\sigma p_0[n+1] - 2g\sqrt{n+1}q_{\mathrm{i}}[n+1]\,, \tag{5a}$$

$$\partial_t p_1[n] = \mathcal{D}_{\mathrm{phot}}\{p_1[n]\} + P_\sigma(T[n] - p_1[n]) + 2g\sqrt{n+1}q_{\mathrm{i}}[n+1]\,, \tag{5b}$$

$$\partial_t q_{\mathrm{i}}[n+1] = \mathcal{D}_{\mathrm{phot}}\{q_{\mathrm{i}}[n+1]\} - \frac{P_\sigma}{2}q_{\mathrm{i}}[n+1] + g\sqrt{n+1}(p_0[n+1] - p_1[n])\,, \tag{5c}$$

where we have separated the photonic dynamics into a superoperator $\mathcal{D}_{\mathrm{phot}}$. Given that it is much slower than the dot dynamics, one can solve the steady state ignoring $\mathcal{D}_{\mathrm{phot}}$ [16]. The photon distribution, $T[n] = p_0[n] + p_1[n]$,

remains unperturbed during the excitation and interaction with the dot. Equations (5) then admit solutions in terms of $T[n]$, i.e., $p_0[n] \approx \frac{\kappa_a(n+1)}{P_\sigma+2\kappa_a(n+1)}T[n]$ and $q_{\rm i}[n] \approx \frac{-2g\sqrt{n}}{P_\sigma+2\kappa_a n}T[n-1]$ where $\kappa_a = 4g^2/P_\sigma$ is the Purcell rate of transfer of population from the cavity to the dot. Our approximations of large intensities imply $n+1 \approx n$.

2. Two-time correlator and spectra — The two-time correlator can be expressed as a sum $\langle\sigma^\dagger(0)\sigma(\tau)\rangle = \sum_{n=0}^{\infty} Q[n](\tau)$, where $Q[n]$ and other functions $S_{0,1}[n]$ and $V[n]$ are defined through the quantum regression formula by coupled differential equations ($n \geq 0$):

$$\partial_\tau Q[n] = \mathcal{D}_{\rm phot}\{Q[n]\} \tag{6a}$$
$$-\frac{P_\sigma}{2}Q[n] + ig(\sqrt{n}S_1[n] - \sqrt{n+1}S_0[n+1])\,,$$
$$\partial_\tau S_0[n+1] = \mathcal{D}_{\rm phot}\{S_0[n+1]\} \tag{6b}$$
$$-P_\sigma S_0[n+1] + ig(\sqrt{n}V[n+1] - \sqrt{n+1}Q[n])\,,$$
$$\partial_\tau S_1[n] = \mathcal{D}_{\rm phot}\{S_1[n]\} \tag{6c}$$
$$+P_\sigma(X[n] - S_1[n]) - ig(\sqrt{n+1}V[n+1] - \sqrt{n}Q[n])\,,$$
$$\partial_\tau V[n+1] = \mathcal{D}_{\rm phot}\{V[n+1]\} \tag{6d}$$
$$-\frac{P_\sigma}{2}V[n+1] + ig(\sqrt{n}S_0[n+1] - \sqrt{n+1}S_1[n])\,.$$

They are, like for the single-time dynamics, separated into a slow photonic dynamics embedded in a superoperator $\mathcal{D}_{\rm phot}$ that is τ-independent in good approximation, and a fast exciton and coupling dynamics that we can solve analytically. Moreover, we have introduced the steady state function $X[n] \equiv S_0[n](0) + S_1[n](0)$, in analogy with $T[n]$. The initial conditions in Eq. (6), are the steady state values $S_0[n+1](0) = iq_{\rm i}[n+1]$, $S_1[n](0) = 0$, $Q[n](0) = p_1[n]$ and $V[n+1](0) = 0$ (therefore, $X[n] = iq_{\rm i}[n]$). After some long, but straightforward algebra, we can find the expression for $Q[n](\tau)$ in terms of $p_{0,1}[n]$ and $q_i[n]$, which, in turn, are expressed in terms of the statistics $T[n]$ (as shown previously). This allows to compute a closed-form solution for $\langle\sigma^\dagger(0)\sigma(\tau)\rangle$, which is however lengthy and not worth writing here. Its main physical features are to reveal that each term in the sum over n, accounts for the 4 possible transitions between the dressed states in the Jaynes-Cummings rungs $n+1$ and n, as in the spontaneous emission case [12]. The first rung, or linear regime, is given by $n = 0$ and consists of only the two transitions of the Rabi doublet. Other rungs give rise to a generalization of the Rabi frequency in the nonlinear regime, the *nth-rung inner* and *outer Rabi frequencies*: $R_{O,I}[n] = \sqrt{g^2(\sqrt{n+1} \pm \sqrt{n})^2 - (P_\sigma/4)^2}$. In the Mollow triplet regime ($P_\sigma > g$), all the peaks positioned at the inner frequencies collapse at the centre (including the Rabi doublet) giving rise to a single central peak. Outer peaks remain split at frequencies $\pm R_O[n] \approx \pm\sqrt{4g^2 n - (P_\sigma/4)^2}$.

The spectrum obtained with the previous derivations can be further simplified for the range of parameters in Eq. (4), to give a compact analytical expression. First, one considers only the coefficients with leading terms in n, making use again of $n+1 \approx n$. Furthermore, due to the Poissonian statistics, only rungs with n close to n_a contribute significantly to the spectra allowing the substitution $n \to n_a$ in $Q[n]$. The sum over n simplifies thanks to the normalization of the distribution function: $\sum_n T[n] = 1$. Finally, we neglect terms related to γ_a before those related to much larger rates, P_σ and κ_σ, i.e., we write the spectrum for these three rates only, through the substitution $g^2 = \kappa_\sigma\gamma_a/4$, and then simply set $\gamma_a \to 0$. This results in the expression for $S_{\rm inc}(\omega)$ in terms of P_σ and κ_σ only:

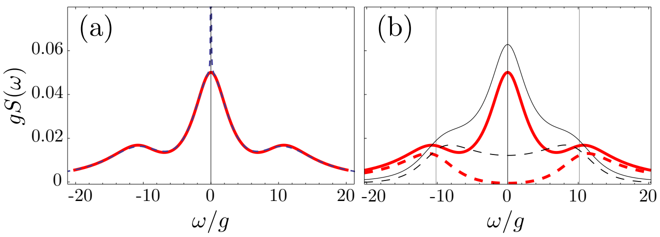

FIG. 2: (Color online) (a) Comparison between our analytical expression Eq. (7), without the elastic peak, (solid red) and the exact numerical solution (dashed blue). (b) Difference between the incoherent (thick solid, red line) and the coherent (thin solid, black line) Mollow triplets, in equivalent conditions. The satellite peaks, that cause the departure, are plotted in dashed. Parameters: $P_\sigma = 6.3g$ and $\gamma_a = 0.1g$.

$$S_{\rm inc}(\omega) = \frac{P_\sigma}{\kappa_\sigma}\frac{\kappa_\sigma - P_\sigma}{\kappa_\sigma + P_\sigma}\delta(\omega) + \frac{1}{2\pi}\frac{\frac{P_\sigma}{2}}{\left(\frac{P_\sigma}{2}\right)^2 + \omega^2}$$
$$+\frac{1}{\pi}\frac{P_\sigma}{\kappa_\sigma + P_\sigma}\frac{(3\kappa_\sigma - P_\sigma)\omega^2 - (\kappa_\sigma - 3P_\sigma)P_\sigma^2}{4\omega^4 - P_\sigma(4\kappa_\sigma - 9P_\sigma)\omega^2 + \kappa_\sigma^2 P_\sigma^2}\,. \tag{7}$$

This is our main result. The structure of the lineshape is the same as that of its coherent counterpart, Eq. (1): a Dirac δ function from the elastically scattered laser light superimposed to a triplet. The central Lorentzian peak has the same weight 1/2 but with FWHM given by the pump, P_σ. The two satellite peaks sit at $\pm\Re(R_O)$ with Mollow splitting:

$$R_O = \frac{P_\sigma}{4}\sqrt{8\kappa_\sigma/P_\sigma - 9} \tag{8}$$

and FWHM $3P_\sigma/2$. The excellent agreement of our formula with exact numerical results [12, 17] is shown in Fig. 2(a), where we superimpose in dashed blue the numerical computation to, in solid red, the analytical expression Eq. (7). Note the elastic peak in the numeric as a very narrow central line.

Despite their similar structure, the lineshapes are intrinsically of a different nature, as can be seen mathematically when reduced to their simplest, dimensionless expression, where they depend only on one parameter

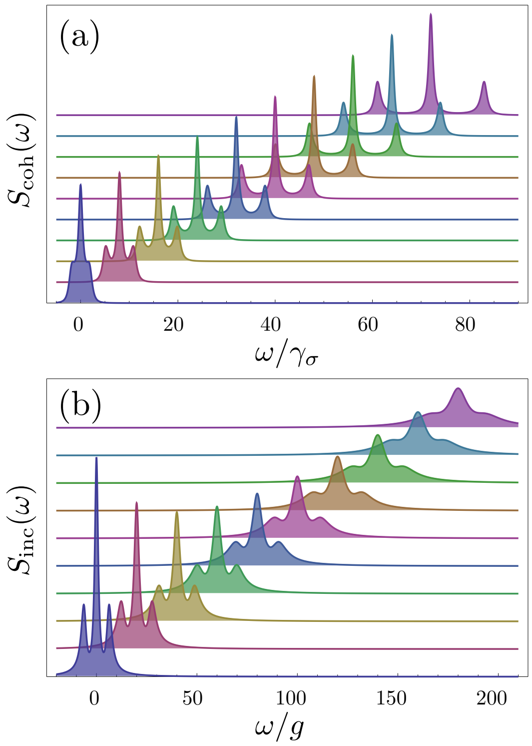

FIG. 3: (Color online) Evolution of the Mollow triplet when increasing (from bottom to top) (a) the coherent excitation (Ω_L/γ_σ from 1 to 5.5 by steps of 0.5) and (b) the incoherent excitation (P_σ/g from 2 to 11 by steps of 1, with $\gamma_a = 0.1g$).

(Ω_L/γ_σ and P_σ/κ_σ). Their similarities and differences can be better appreciated when compared on physical grounds, when the laser intensity for the conventional Mollow triplet, Eq. (1), is taken the same as the average population of the cavity under incoherent excitation (i.e., we take $\Omega_L^2 \to g^2 n_a \approx P_\sigma(\kappa_\sigma - P_\sigma)/8$), and when the broadening of the dot includes P_σ. In this way, we attempt the description of the incoherent system with the theory of the coherent one. We obtain an expression that shows the fundamental discrepancies between the two types of triplets:

$$S_{\rm coh}(\omega) = \frac{P_\sigma}{\kappa_\sigma}\delta(\omega) + \frac{1}{2\pi}\frac{\frac{P_\sigma}{2}}{\left(\frac{P_\sigma}{2}\right)^2 + \omega^2} - \frac{1}{\pi}\frac{P_\sigma\omega^2 - (2\kappa_\sigma - 3P_\sigma)P_\sigma^2}{4\omega^4 - P_\sigma(4\kappa_\sigma - 9P_\sigma)\omega^2 + \kappa_\sigma^2 P_\sigma^2}\,. \quad (9)$$

Comparing this expression with Eq. (7), one can see, indeed, that the central peak is the same in both cases, as well as the position and broadening of the satellite peaks (third terms have the same denominator), so the underlying structures bear much similarities. However the satellite lineshapes differ considerably, being strongly affected by the effect of incoherent pumping, that renormalizes the rungs of the Jaynes-Cummings ladder, and other factors such as the dot population, which under coherent excitation shows opposite behaviour to that of Eq. (3): $n_\sigma^{\rm coh} \approx \frac{1}{2}(1 - P_\sigma/\kappa_\sigma)$. The shapes of these peaks are shown in more details in Fig. 2(b), where they are plotted (in dashed) together with the whole triplets, in the coherent (thin black) and the incoherent (thick red) cases.

Finally, Fig. 3 shows the natural experimental configuration to demonstrate the new character of nonlinear spectroscopy in microcavities under incoherent pumping, and to contrast it with its coherent counterpart. Increasing pumping, one sees that in the coherent case (upper panel), the triplet is better resolved, with a larger splitting, while in the incoherent case (lower panel), the satellites overlap with the central line as a result from pumping that splits them sublinearly, Eq. (8), and also increases their broadening. The two phenomenologies, despite their deep interconnections and common features, are strikingly different and the evidence of the new one should pose no problem even on qualitative grounds.

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